IJCRT.ORG ISSN : 2320-2882

INTERNATIONAL JOURNAL OF CREATIVE RESEARCH THOUGHTS (IJCRT)

An International Open Access, Peer-reviewed, Refereed Journal

HEALING BEHAVIOUR OF CARBON FIBER REINFORCED POLYMER COMPOSITES

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Abstract: This research paper investigates the healing performance of epoxy matrix carbon fiber reinforced polymer composites, focusing on their ability to self-repair structural damage. The study explores the potential of these composites to autonomously heal microcracks and delamination, which can occur during service conditions, thereby extending the operational lifespan of composite structures. Through a comprehensive literature review, various healing mechanisms in composite materials are examined, highlighting the significance of healing performance in enhancing material durability and reliability. Experimental methods are employed to assess the healing capabilities of the composites, including characterization techniques such as optical microscopy, mechanical testing. This test are performed by varying the GO content from 0% to 1%. Thus maximum amount of fracture load obtained at the 0.50%.The results provide insights into the effectiveness of different healing mechanisms and their influence on the mechanical properties of the composites. The findings of this study contribute to the understanding of self-healing materials and have implications for the development of advanced composite structures with improved damage tolerance and longevity.

Index Terms **– Epoxy, Carbon fiber, Graphene oxide, catalyst, peel ply.**

I. INTRODUCTION

Carbon fiber reinforced composites have emerged as prominent materials in various engineering sectors, particularly in aerospace applications, owing to their exceptional specific strength and lightweight properties. These composites offer significant advantages over traditional materials, providing enhanced performance and efficiency in demanding environments. However, despite their strengths, carbon fiber reinforced composites are susceptible to damage from relatively low-velocity and localized impact loads, particularly in the through-thickness direction. The mechanical stresses imposed during service conditions can induce structural damage, leading to delamination—characterized by separation cracks—at the interfaces between fiber layers. Delamination not only compromises the structural integrity of the composite but also undermines its overall performance and reliability [1]. Therefore, addressing the challenges associated with delamination and enhancing the damage tolerance of carbon fiber reinforced composites is imperative to ensure their sustained use in critical applications.

Various techniques have been devised to bolster the resistance of composites against delamination, with a focus on enhancing interlaminar strength without compromising in-plane mechanical properties [2]. One approach involves modifying the polymer matrix with nanofillers, such as multiwalled carbon nanotubes (CNTs), fullerenes, and nano clay, which have shown promising results in augmenting the shear strength, flexural strength, and fracture toughness of composite materials [3]. Thakre et al. demonstrated that a synergistic combination of pristine and functionalized single-walled CNTs (SWNTs) enhanced the interlaminar properties of carbon fiber composites [4]. Similarly, Wicks et al. achieved a remarkable 76% improvement in interlaminar fracture toughness by incorporating aligned CNTs into the interlaminar area of fiberglass–epoxy composites [5]. Through chemical vapor deposition, Garcia et al. successfully increased interlaminar fracture toughness by approximately 150% in carbon fiber–epoxy prepreg-based composites [6]. Arai et al. observed a 50% improvement in Mode I interlaminar fracture toughness by introducing vapor-grown carbon fibers (VGCFs) and carbon nanofibers (CNFs) into the interlaminar region of carbon fiber–epoxy laminates [7]. Additionally, an interlayer comprising functionalized XD CNT/epoxy demonstrated an 18% enhancement in initial fracture toughness over standard panels and a 36% improvement over panels with only epoxy as an interlayer [8]. These advancements underscore the potential of nanofiller modification to bolster interlaminar bonding and mitigate delamination in composite structures, thus aligning with the objectives of this research paper [5].

Recent advances in materials science have sparked a keen interest in incorporating graphene and graphene oxide (GO) as potential additives for composite materials [9], [10]. Of particular note is GO, with its distinctive physical properties and nanoscale dimensions, making it an appealing candidate for reinforcing fillers in polymer composites, thanks to its amphiphilic nature [11], [12], [13]. Despite its potential, there is a gap in exploring GO's role in interlaminar modification of carbon fiber reinforced composites. The effective dispersion of nanofillers within the polymer matrix is paramount for enhancing mechanical properties, a challenge addressed through various dispersion methods such as sonication, shear mixing, and the use of compatibilizers like polyvinylpyrrolidone (PVP) [14], [15]. PVP, renowned for its exceptional crosslinking properties and compatibility with diverse

matrix resins, plays a pivotal role in dispersing and stabilizing nanoparticles within the interlaminar region, thereby enhancing fiber-matrix bonding and overall mechanical properties [16], [17]. Furthermore, PVP shows promise as an intercalant for plate-like nanoparticles, offering further insights into its potential to enhance composite performance [18], [19]. This review underscores the significance of incorporating GO and utilizing PVP as a compatibilizer to improve the mechanical properties and interlaminar bonding of carbon fiber reinforced composites, aligning seamlessly with the objectives of this research paper.

In this research, the use of graphene oxide (GO) is explored to enhance the mechanical properties and interlaminar bonding of carbon fiber reinforced composites. By optimizing the dispersion of GO within the epoxy matrix using various techniques, including sonication and the addition of polyvinylpyrrolidone (PVP) as a compatibilizer, the aim is to improve the fracture load and overall mechanical performance of the composites. Through experimental methods such as mechanical testing and optical microscopy, the effectiveness of GO in reinforcing the composites is assessed, along with potential synergies with PVP. This study contributes to the development of self-healing composite materials with enhanced durability and reliability.

II. MATERIALS AND METHODOLOGY

2.1. Materials:

A bidirectional Carbon Fiber (CF) mat with a GSM 200 and filament diameter of 7 µm was purchased from Bhor Chemicals. Diglycidyl ether of bisphenol A (DGEBA) epoxy prepolymer with an amine-based hardener system was purchased from Atul Industries. Ethanol (C_2H_5OH) and doubled Distilled Deionized water were purchased from Merck/Sigma-Aldrich. Graphene oxide (GO) was obtained from Nano research Laboratory (NRL), Hyderabad. In addition to that the following consumable materials are required to prepare the composites samples for evaluating fracture toughness and the healing efficiency of composites. This includes: Carbon Fiber (400 GSM), Graphene nano Powder, Epoxy Resin and Hardener, Steel Plates Mold, and Teflon tape.

2.2. Equipments:

In the present research work, the following equipments are required to prepare the composites samples for evaluating fracture toughness and the healing efficiency of composites. This includes: Mini Overhead Stirrer, Vacuum Desiccator, Vacuum Pump, Hand Lay-Up Setup and Steel Mold for preparation of FRP plates as shown in Fig. II-1 to II-4.

Figure II-1. Remi Mini Stirrer RQ-5Plus

Figure II-2. Vacuum Desiccator

Figure II-3. Operating Vacuum Desiccator with Vacuum Pump

Figure II-4. Hand Lay-Up set up

2.3. Methodology:

The methodology employed in this study follows a systematic approach outlined in 5 below. Each step in the fabrication process is meticulously executed to ensure the production of high-quality composite specimens for subsequent testing and analysis. This methodology outlines the steps involved in preparing, molding, post-curing, testing, and analyzing the double cantilever beam specimens made of carbon fiber reinforced composite with graphene nanoparticles, providing a comprehensive approach to investigate its mechanical properties and interlaminar fracture toughness.

In the Materials and Method section, the carbon fiber utilized in this study is a 400 GSM (grams per square meter) material known for its exceptional strength and stiffness, making it ideal for demanding applications in aerospace, automotive, and sporting goods industries. This biaxial carbon fiber comprises eight layers of fabric, each weighing 200 g/m^2 offering a balance between strength and weight. Suitable for hand lay-up, infusion, or RTM (resin transfer molding) processes, it is compatible with epoxy, urethane-acrylate, or vinyl ester resin systems. Additionally, peel ply with a distinctive red stripe, weighing 85 gpsm, serves as an effective porous release fabric. Stabilized at high temperatures, it facilitates resin infusion or hand lay-up processes by allowing excess resin and air to escape, minimizing voids in the composite parts. The peel ply, free of release agents or contaminants, can be directly applied to the part surface and easily removed after curing, reducing the need for post-processing. Furthermore, the laboratory stirrer used for epoxy mixture preparation is the RQ-5 Plus, equipped with advanced features such as high torque even at low speeds, stirring speed control through VFD (variable frequency drive), and motor protection against overload. With a brushless DC motor rating of 10 watts, the stirrer operates within a speed range of 50 to 1500 rpm and offers precise control over stirring time, making it an essential tool for ensuring the uniform mixing of epoxy resin and hardener.

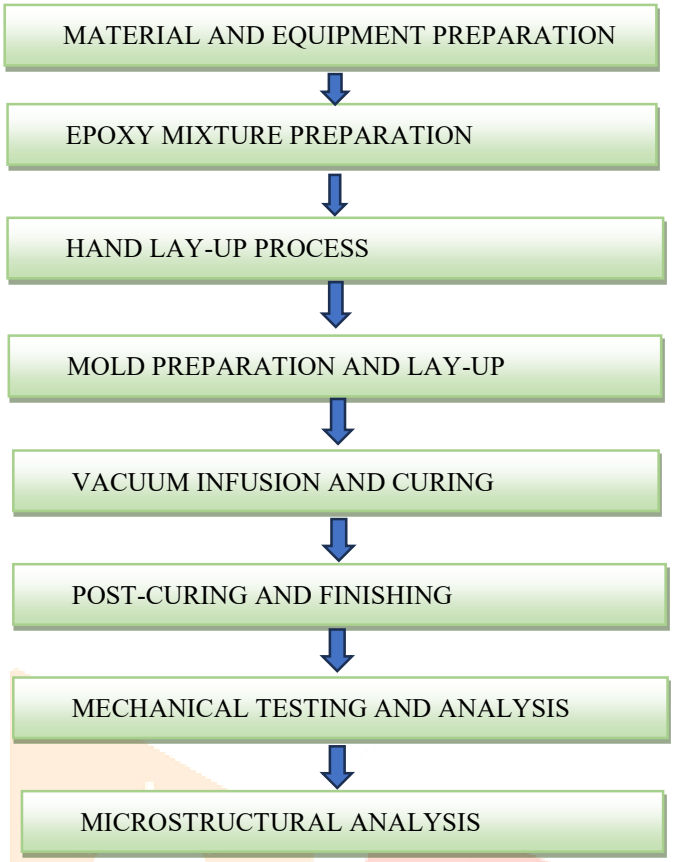

Figure II-5. Process flow chart for methodology of present research work

2.4 Epoxy Mixture Preparation

In the Epoxy Mixture Preparation section, Lapox K-6, a light-yellow aliphatic polyamine hardener, is carefully combined with epoxy resin (L12) in the appropriate ratio of 10:1 to form the epoxy mixture. Lapox K-6 is specifically designed to cure epoxy resin at ambient temperatures, offering short pot life and faster cure speed due to its very low viscosity and low dosage. This makes it suitable for various applications such as adhesives, mortar, and casting of small electrical components. The epoxy resin, LY556, along with the K6 hardener, is commonly utilized in composite material applications for its superior bonding properties and durability. Graphene oxide (GO) is another crucial component incorporated into the epoxy mixture composition. GO contains approximately 77% carbon and 22% oxygen, with trace amounts of other elements, resulting in a carbon purity of approximately 99% and a bulk density of 0.48g/cc. GO nanoparticles exhibit an excellent aspect ratio, with lengths reaching 5 to 10m, and possess a very high surface area advantageous for adsorption and surface interaction. They also act as effective barriers against gases and liquids and offer tensile strength many times greater than steel, making them highly durable and suitable for various applications. Additionally, GO nanoparticles have the potential to contribute to sustainability efforts due to their lightweight nature.

The composition preparation process involves carefully measuring and mixing the epoxy resin and graphene oxide in varying proportions using electronic composite scales. The resin and hardener are combined in the appropriate ratio, while GO powder is added in percentages ranging from 0.25% to 1%. After manual mixing, the mixture is further stirred using a Remi mini stirrer at a constant speed for 1-2 hours to ensure thorough blending. Subsequently, the mixture is placed in a vacuum desiccator operated by a vacuum pump for 2-3 hours to eliminate air bubbles, thereby reducing defects and improving the strength and stiffness of the composite material. This meticulous preparation of the epoxy mixture, along with the controlled incorporation of graphene oxide, is essential for achieving consistent mechanical properties and ensuring the overall quality and reliability of the composite materials.

2.5 Hand Lay-Up Process

During the hand lay-up process, carbon fiber prepreg sheets are manually laid onto the mold or template in a precise arrangement. This involves carefully positioning the sheets to ensure proper fiber orientation and alignment according to the desired specifications for the Double Cantilever Beam (DCB) specimen. Additionally, epoxy resin mixture is applied between the layers to impregnate the carbon fibers and create a strong bond between them. This manual process allows for flexibility in adjusting the lay-up to accommodate specific design requirements or variations in material properties. Attention to detail during hand lay-up is crucial to achieving uniform resin distribution and fiber alignment, which ultimately contribute to the mechanical performance of the final composite structure.

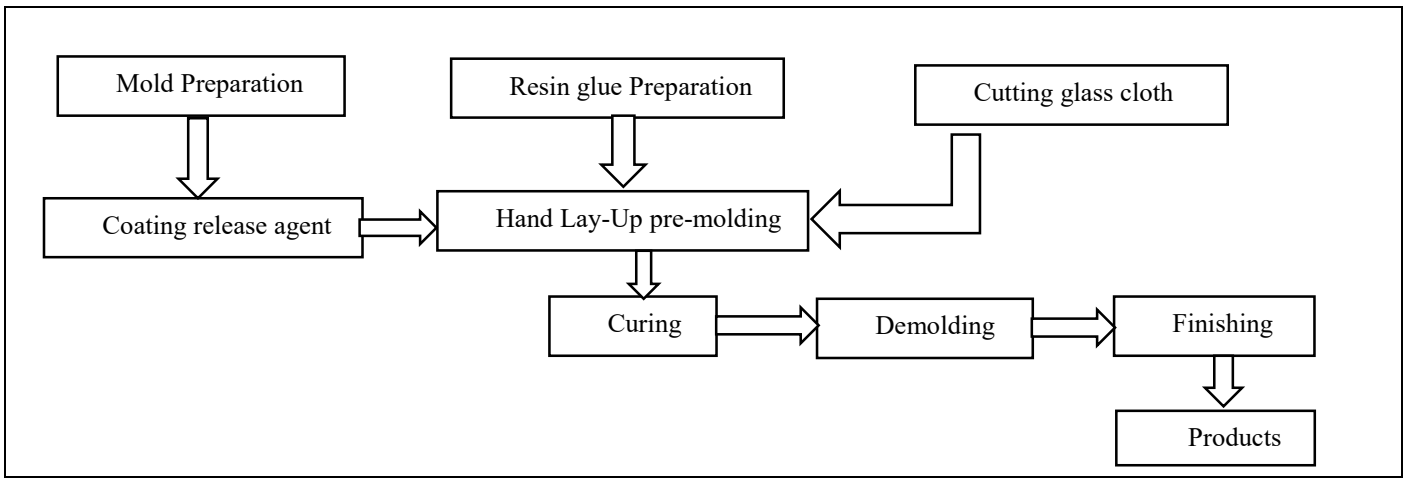

Figure II-6 Technic Flowchart for FRP by Hand Lay-Up

2.6 Mold Preparation and Lay-Up

In the mold preparation and lay-up stage, the mold or template is first treated with a release agent to prevent the carbon fiber composite from sticking during curing. Thus, the six layers of carbon fiber sheet is used during preparation. Between the three layers of This sheets Teflon is placed from one end of the DCB Plates.to ensures easy removal of the finished Double Cantilever Beam (DCB) specimen from the mold once the lay-up is complete. Next, the carbon fiber prepreg sheets are carefully arranged and laid onto the prepared mold in accordance with the desired configuration for the DCB specimen. Proper alignment and orientation of the carbon fibers are crucial to achieving optimal mechanical properties in the final composite structure. The lay-up process may involve multiple layers of prepreg sheets, with epoxy resin applied between each layer to facilitate bonding and impregnation of the fibers. Attention to detail during mold preparation and lay-up is essential for producing high-quality DCB specimens with consistent material properties and performance characteristics.

2.7 Vacuum Infusion and Curing

During vacuum infusion and curing, the prepared lay-up of carbon fiber prepreg sheets and epoxy resin is subjected to a vacuum bagging process to ensure thorough impregnation of the fibers and consolidation of the composite structure. First, vacuum bagging materials such as vacuum bag film, breather cloth, and sealant tape are applied to create a sealed enclosure around the lay-up. This vacuum-tight seal is essential for creating a controlled environment during the infusion process. Once the vacuum bagging materials are in place, a vacuum pump is used to evacuate the air from within the enclosure, creating a negative pressure environment. This vacuum pressure helps to remove trapped air and excess resin from the lay-up, promoting resin flow and uniform distribution throughout the composite. Next, the lay-up is infused with epoxy resin under vacuum pressure. The resin is introduced into the lay-up either through injection ports or by manually spreading it over the surface of the lay-up. The vacuum pressure helps to draw the resin into the carbon fiber layers, ensuring thorough wetting and impregnation of the fibers. After the infusion process is complete, the vacuum bagged lay-up is transferred to a curing oven or autoclave for thermal curing. The curing cycle involves subjecting the composite to elevated temperature and pressure conditions, typically in accordance with the resin manufacturer's specifications. This curing process activates the epoxy resin, causing it to cross-link and bond the carbon fibers together, resulting in a fully cured and consolidated composite structure. Once cured, the vacuum bagging materials are removed, and the finished Double Cantilever Beam (DCB) specimen is extracted from the mold. The cured specimen may undergo additional post-curing steps or finishing processes as needed before mechanical testing and analysis. Overall, vacuum infusion and curing are critical steps in the fabrication of high-quality carbon fiber composites, ensuring optimal resin distribution and consolidation for superior mechanical performance.

2.8 Post-Curing and Drying

After the initial curing process, the Double Cantilever Beam (DCB) Plates may undergo post-curing to further enhance its mechanical properties. Post-curing involves subjecting the cured composite to additional time and temperature cycles under controlled conditions. This helps to optimize the cross-linking of the epoxy resin and improve the overall strength, stiffness, and durability of the composite material. Once the post-curing process is complete, the DCB specimen may require finishing to achieve the desired surface quality and dimensions. Finishing techniques may include sanding, machining, or polishing to remove any rough edges, excess resin, or imperfections from the surface of the specimen. This ensures a smooth and uniform finish that meets the specified dimensional tolerances for testing and analysis. Additionally, any necessary markings or identification may be applied to the specimen during the finishing process for traceability and documentation purpose. After the marking cutting the specimens into (ASTM D5528) standards. This could include labeling the specimen with relevant information such as material type, lay-up configuration, curing parameters, and test specimen identification numbers.

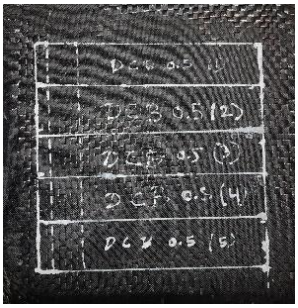

Figure II-1 Sample Panel 1

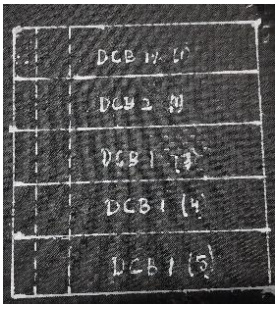

Figure II-2 Sample Panel 2

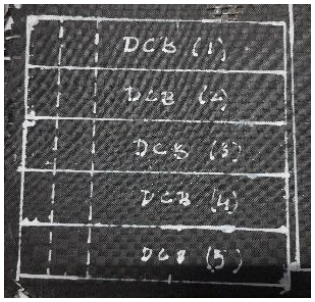

Figure II-3 Sample Panel 3

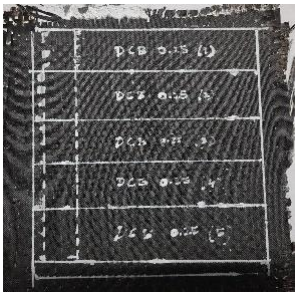

Figure II-4 Sample Panel 4

Table II-1. Composition for preparing CFRP Plate

Sample panels	Wt (%) GO	Epoxy(L12) Content	Graphene oxide (GO) Content	Hardner (K6) content
1	0	200 gm	0	20 gm
2	0.25	170 gm	0.45 gm	17 gm
3	0.5	120 gm	0.6 gm	12 gm
4	1	170 gm	1.7 gm	17 gm

III. Mechanical Testing and Microstructural analysis

3.1 Mechanical Testing:

Once the Double Cantilever Beam (DCB) specimen is fabricated and finished, it undergoes mechanical testing to evaluate its performance characteristics, particularly its interlaminar fracture toughness. The DCB test typically involves applying a controlled displacement or load to the specimen, causing a crack to propagate along the interface between the laminates. This test measures the energy required to propagate the crack and provides valuable information about the material's resistance to interlaminar fracture. Mechanical testing is conducted using a universal testing machine equipped with appropriate grips and fixtures designed for DCB testing. The specimen is securely clamped in the grips, and a load is applied at a constant rate or displacement rate until the crack propagates across the specimen. During testing, data such as load-displacement curves and crack growth behavior are recorded and analyzed to determine key parameters such as critical load, critical displacement, and fracture toughness. These parameters help assess the material's ability to withstand crack propagation and resist interlaminar delamination. Following mechanical testing, detailed analysis of the test results is performed to understand the material's behavior under loading conditions and identify any failure mechanisms or deficiencies. This may involve comparing the experimental data with theoretical models or numerical simulations to validate the material's performance and predict its behavior in real-world applications.

Figure II-5. DCB Specimen

Figure II-6. DCB Test on UTM

3.2 Microstructural analysis:

Microstructural analysis involves examining the internal structure of the carbon fiber composite at a microscopic level to understand its composition, morphology, and integrity. This analysis provides insights into the distribution of fibers, resin matrix, voids, and any defects or anomalies present within the material.

3.2.1 Scanning Electron Microscopy (SEM):

Scanning Electron Microscopy (SEM) is a powerful technique used to examine the microstructure of materials at high resolution. In the context of carbon fiber composites, SEM provides detailed images of the composite's surface and cross-section, revealing information about fiber morphology, resin distribution, fiber-matrix interfaces, and the presence of defects or damage. During SEM analysis, the specimen is placed in a vacuum chamber and bombarded with a focused beam of electrons. The interaction of the electrons with the atoms in the specimen produces signals such as secondary electrons, backscattered electrons, and X-rays. These signals are detected and used to generate images that reveal the topography, composition, and other properties of the specimen at the microscale.

3.2.2 Failure mode of Layer composites:

SEM can provide valuable insights into the following aspects of carbon fiber composites:

- Fiber Morphology:

SEM images can reveal the size, shape, and alignment of individual carbon fibers within the composite. This information is critical for assessing the quality and integrity of the fibers and understanding how they contribute to the mechanical properties of the material.

- Resin Distribution:

SEM can show how the resin matrix is distributed within the composite, including how it fills the spaces between fibers and any voids or defects present. This helps assess the degree of resin impregnation and uniformity in the composite, which is important for achieving optimal mechanical performance.

- Fiber-Matrix Interfaces:

SEM images can highlight the interface between carbon fibers and the resin matrix, providing insights into the adhesion and bonding between the two phases. A strong interface is essential for transferring loads between fibers and the matrix and maximizing the composite's strength and stiffness.

- Defects and Damage:

SEM can detect and characterize defects such as voids, delamination, cracks, and fiber pull-out within the composite. Understanding the nature and extent of these defects helps identify potential sources of weakness or failure and informs strategies for improving the fabrication process and enhancing material performance.

3.2.3 Healing efficiency calculations:

The calculations of the healing efficiency (H.E.) of the estimated system were based on Equation (1):

$$H.E = \frac{P\ max_{healed}}{P\ max_{before\ healed}} = \frac{G_{healed}}{G_{before\ healed}} \text{ - - - - - - - - - - (1)}$$

Where, H.E = healing efficiency, Pmax = peak Load, G = Fracture toughness energy

IV. RESULTS AND DISCUSSION

4.1 DCB Fracture Test:

The fracture toughness property of the hierarchical laminates was observed by the performance test of the Double cantilever Beam (DCB). The samples of epoxy laminates composites was prepared with 0 wt %, 0.25 wt %, 0.5 wt % GO and 1 wt % GO, respectively. In all laminated composites, GO was uniformly distributed in all 8 layers of CF lamina. Similarly, neat epoxy CFRP composites were also prepared without using GO nanopowder in all of the 10 layers of CF mats. The laminates were fabricated by the Hand layup technique followed by compression molding.

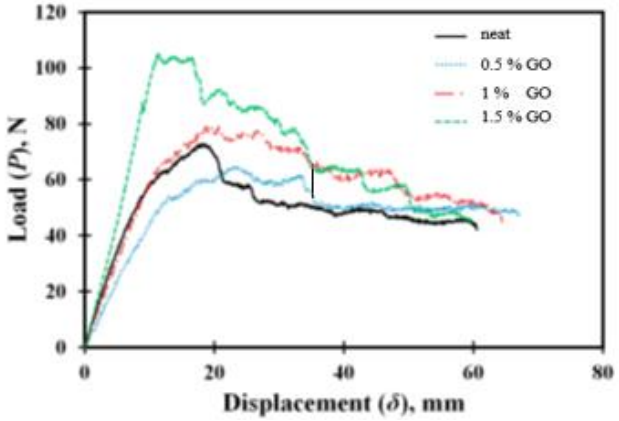

Figure IV-1 the load-displacement curves for DCB specimens with three different contents of GO

Figure IV-1 depicts a comparison of the load-displacement curves between the three tested GO contents. The maximum load and stiffness of the 0.5 wt% GO composite specimen displayed an increase compared to the other three specimen types. The maximum load and stiffness of the composites with 0.25 wt% GO content specimen remained about the same as the neat specimen, while the 1.0 wt% GO content specimen dropped slightly in terms of maximum load and stiffness.

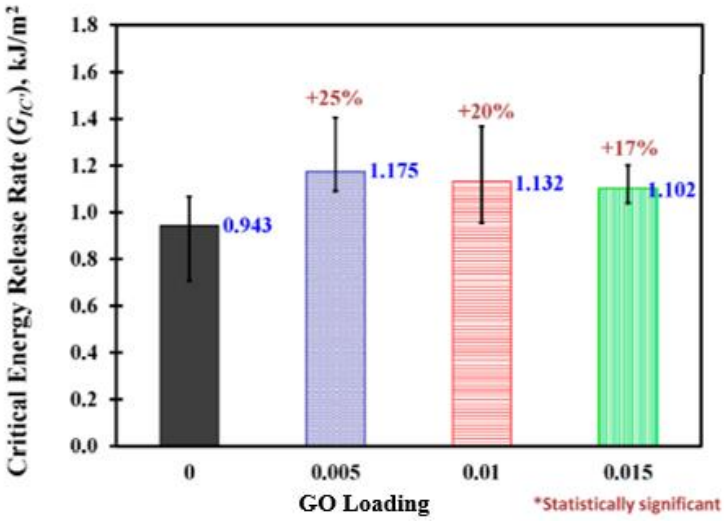

Figure IV-2 A comparison of the maximum critical energy release rates for DCB specimens with three different contents of GO

It is believed that in neat CFRP laminates, delamination occurs due to tensile cracking of the matrix associated with bridging of carbon fibers, which provides resistance against delamination growth at the crack tip. When GO are added, a similar mechanism remains; however, an additional energy consumption mechanism is developed where the GO also bridge the interface of the crack tip and provide increased resistance against crack propagation. An LED illuminated digital optical microscope was used to examine the fracture surface morphology of specimens containing 0.0, 0.25, 0.5, and 1.0 wt% GO, displayed in Figure IV-2. The surface of the neat specimen displays relatively smooth epoxy. The remaining three specimens show evidence of GO pull-out and fiber bridging observed in their rough fracture surface. Li et al. [15] confirmed that composites containing CNTs produced rougher fracture surfaces compared with composites without GO. This is likely due to a toughening effect taking place in the matrix. Toughening can hinder crack propagation by reducing stress concentrations and producing a strong interphase.

4.2 Self-Healing Properties:

The failure mode for the DCB samples was through the "Tensile mode". Therefore, the toughness recovery analysis was performed on the DCB samples of epoxy matrix CF with 0.5 wt % GO composites. The failed DCB samples were kept in an oven at 60 °C for 24 h, followed by oven cooling for self-healing. The samples were tested again in the UTM to gauge the recovery of toughness post failure. Figure IV-3 (a) shows the maximum load (Pmax) and mode II interlaminar fracture toughness energy (G_{IIC}) values of the hybrid CFRP, before and after the healing cycles, and Figure IV-3 (b) shows The effect of the number of healing cycles on the healing efficiency of epoxy matrix CF with 0.5 wt % GO composites.

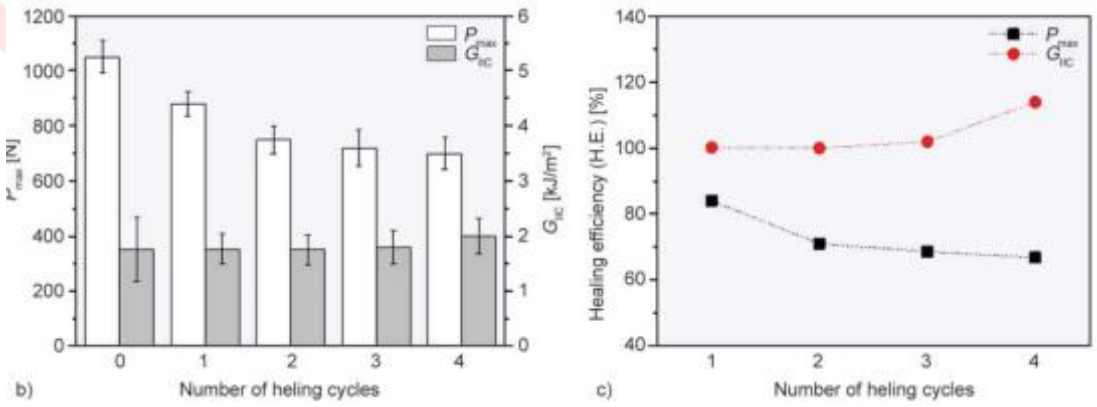

Figure IV-3 (a) shows the maximum load (Pmax) and mode II interlaminar fracture toughness energy (G_{IIC}) values of hybrid CFRP, before and after the healing cycles, and Figure IV-3 (b) shows the effect of the number of healing cycles on the healing efficiency of epoxy matrix CF with 0.5 wt % GO composites.

In the histogram of Figure IV-3 (a) the effect of the number of healing cycles on the mode II fracture characteristics (Pmax and G_{IIC}) of the hybrid CFRP is seen. As for the Pmax, the four healing cycles were able to reduce the recorded Pmax value at a rate of approximately 53%. After the first healing activation the Pmax of the hybrid CFRP showed a decrease of approximately 22%. The same behavior was observed after the second healing cycle when compared to the first one. Following the third healing cycle the decrease of the Pmax was calculated to be close to10 % comparing to the second one while the same decrease proportion was calculated after the fourth healing event. On the other hand the G_{IIC} values exhibit a slight increase with increasing the number of healing activations; the G_{IIC} value after the fourth healing cycle is 14% higher than that in the original (no-healed) situation. This behavior is attributed to the fact that the displacement value at crack propagation onset increases as the healing cycles increase (see Equation (1)). In Figure IV-3 (b) the actual Healing Efficiency (H.E.) values for the two magnitudes of interest, calculated based on Equation (2), are given as a function of the number of healing cycles. In subsequent healing cycles a drop for the H. E. and Pmax is observed. Nevertheless the lowest value still remains above 70% of the reference value.

V. CONCLUSIONS

In this work, the fracture toughness and healing efficiency of GO reinforced epoxy matrix layered composites was investigated. Mode II interlaminar fracture toughness tests were performed to assess the effects the healing agent on healing efficiency of composites. It was shown that the healing agent epoxy resin which was introduced at the mid-plane of the lay-up notably increased the fracture toughness of the hybrid composite system, when compared to the reference one. The Pmax doubled while the GIIC multiplied two times. This has been justified by the effective recovery of more than 85% of the maximum load and 100% of the fracture toughness of the hybrid composite. A decreasing healing ability was observed in terms of load bearing capacity.

VI. ACKNOWLEDGMENT

This research work was partially completed specially all the samples were prepared at Godavari Institute of Engineering and technology, Rajahmundry, and the DCB test and SEM characterization at VIRTUE META-SOL, Hyderabad. The authors acknowledge both supports. The third author greatly acknowledges Literature survey. All the authors would also like to thank Dr. Tankeshwar Prasad for his initial help and guidance in conducting the experiments, Mr. Kiran Kumar for his assistance in conducting the Mechanical Testing and SEM characterizations.

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